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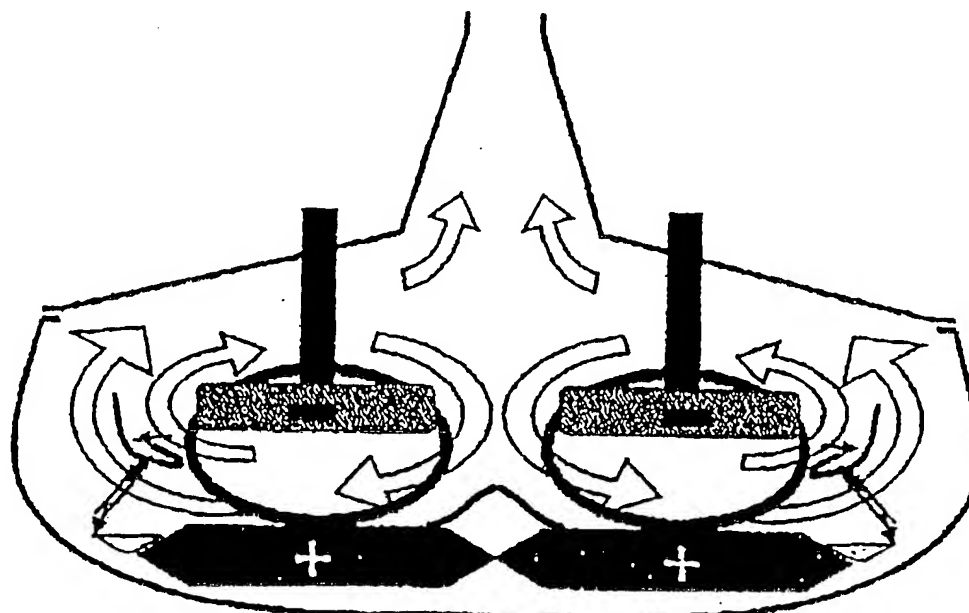
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(54) Title: ELECTROLYTIC CELLS SWEEP BY AN ELECTROMAGNETIC FIELD AND PROCESS THEREFOR

(57) Abstract

The present invention relates to a molten electrolyte electrolysis apparatus and process and their embodiments which are adapted for the reduction of metallic minerals. The device according to the present invention can be adapted for the production of the principle industrial metals such as zinc, magnesium, aluminium, titanium, etc. An electrolytic cell comprising electrolyte, at least one anode and at least one cathode, characterized in that electrolyte is caused to circulate between the cathode(s) and anode(s) by imposition of a magnetic field typically perpendicular to the passage of current in the electrolyte and

associated with one or several deflectors which separate the electrolyte flow into two flow paths, the first flow path passing the anode and the second flow path passing the cathode. According to the present invention, the cells can be adapted for the treatment of ores with the separation of different metals inside.



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ELECTROLYTIC CELLS SWEEPED BY AN ELECTROMAGNETIC FIELD AND PROCESS THEREFOR

The present invention relates to a molten electrolyte electrolysis apparatus and process and their embodiments which are adapted for the reduction of metallic minerals. The device according to the present invention can be adapted for the production of the principle industrial metal such as zinc, magnesium, aluminium, titanium, and also zirconium, tungsten, lead, etcetera. Certain of the embodiments of the invention can allow the simultaneous separation of different metals which are common constituents of ores, therefore producing an improved economy of production which is capable of competing favorably with the traditional industrial technology of the prior art.

The principle ores processed to obtain metals generally comprise oxides, sulfates or carbonates. By roasting, the sulfates are transformed to oxides while the carbonates generally undergo a decarbonation which also converts them to oxides. It is the reduction of these oxides which generally produces the pure metal. This reduction of oxides can be achieved either by thermo-metallic reaction or by electrolysis.

Thermo-metallic reaction comprises displacing oxygen from an oxide by another, more reductive metal, or fixation by carbon with production of carbon containing gas, in which case the process is called a thermo-carbonic reaction. Electrolysis causes the decomposition of a molecule. In this case the oxide is often transformed into chloride, such as, for example in the electrolysis of magnesium or zinc. Alternatively, the oxide is put in solution in a molten salt bath, such as in case of aluminium oxide which is put in solution in a molten cryolite at high temperature. In an electrolysis cell, the metal deposits on the cathode while the oxygen or halogen is released at the anode and can then participate in the reaction with the anode.

The industry has a particular type of treatment cells adapted for the reduction of each type of oxide. Although each metal corresponds to a particular type of cell, one can nevertheless classify the electrolytic cells according to their different types.

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To begin with, one can categorise the electrolytic cells into multi-electrode cells and into mono-cathode cells.

In the multi-electrodes cells, the electrodes are generally placed vertically, alternating cathodes with anodes, while in the mono-cathode cells, the bottom of the bath comprises the electrode and the bottom is necessarily in a horizontal orientation.

For example the electrolysis of zinc chloride in aqueous media is said to use a low temperature multi electrode cell, and the zinc is fixed directly on the cathodes. Likewise, for the electrolysis of molten magnesium chloride, a multi electrodes cell is used. In this case, at the temperature of electrolysis, the magnesium is liquid and being less dense, it goes up the length of the cathodes and floats at the surface of the bath. If diaphragms are not interposed between the anodes and cathodes, the chloride which is released from the anodes recombines with the magnesium and the cell efficiency is null.

As distinct from the preceding electrolysis cells, mono-cathodic electrolysis cells comprise only one cathode which makes up the bottom of the cell, while one or more anodes are immersed in the electrolyte above the cathode. One of the best known mono-cathodic cell is the aluminium electrolysis cell. This apparatus is particularly well adapted for the electrolysis of aluminium at the elevated temperatures of electrolysis; the density of aluminium is slightly greater than the density of the electrolyte in the cell, so that aluminium can accumulate at the bottom of the bath while the oxygen liberated at the upper anodes forms carbon-bearing gas, without any risk of recombining with the metal deposited at the bottom of the bath.

A priori, the arrangement comprising an anode at the bottom of the bath and one or more cathodes in an upper position, that is to say, an inversed arrangement, would not be possible. The problem is that when a metal is of lower density than the density of the electrolyte, the material released at the anode inevitably become recombined with the metal.

According to another method of classification, it is also possible to distinguish the electrolysis cells according to whether the electrolysis is carried

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out at low temperatures in an aqueous medium, or carried at high temperature in a bath of molten salt.

Alternatively, it is also possible to classify the cells according to the nature of the electrolyte or whether or not it is in circulation. But this last
5 classification can overlap with preceding classifications.

In effect, during electrolysis in an aqueous media, the electrolyte may be put into continuous circulation with the aid of pumps, such that the (titrage) baths are controlled, filtered and regulated. Furthermore the sweeping or flowing movement of the electrolyte assists the anodic depolarization and
10 improves the deposition at the cathode.

By contrast, in the electrolytic cells (ignee) or molten salt baths, the aggressiveness of the electrolyte at high temperature makes it virtually impossible to put the electrolyte into circulation using mechanical pumps centrifuges or other pumps. Moreover, it is undesirable to have excessive
15 perturbation of the electrolyte baths, particularly molten salt baths, and generally, there is no circulation of electrolyte in electrolytic cells having a molten electrolyte. Thus, for example, in the electrolysis of aluminium, the densities of the different components of the bath are very similar, and agitation of the bath can cause collisions between the cathode and anodes. These
20 collisions can then cause destructive electromagnetic reactions.

It is known that a magnetic field can be used as the source of movement of electrolyte of an electrolysis cell, but the problem is that in reality, the magnetic fields caused by the passage of current in industrial electrolytic cells, do not have perceptible effects. With respect to multi-electrode cells, the
25 magnetic fields are generally compensated for or negated by other effects, while in the mono-cathodic cells such as aluminium electrolysis cells, the passage of current in the electrolyte causes a vertical, axially centred magnetic pressure and has little effect (or causes stabilizing effects) and are incapable of causing a circulation of electrolyte between the electrodes.

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It has now been found, that a device and process can be provided in which there is forced circulation of the electrolyte between anodes and cathodes of an electrolytic cell.

The present invention therefor provides an electromagnetic cell comprising electrolyte, at least one anode and at least one cathode, characterized in that electrolyte is caused to circulate between the cathode(s) and anode(s) by imposition of a magnetic field. Typically the magnetic field is perpendicular to the passage of current in the electrolyte. Typically deflectors are included in the apparatus to direct the flow of electrolyte.

The present invention also provides a method of circulating electrolyte between the cathode(s) and anode(s) of an electrolytic cell comprising the step of imposing a magnetic field. Furthermore, the present invention also provides unique methods of processing metals, metallic minerals and the like using the apparatus and process of the present invention.

One benefit or effect resulting from the forced circulation of electrolyte or sweeping of electrodes is an improvement of the efficiency of the electrolytic cell due to depolarisation of the anode. A further benefit of effect is a reduction in wear of the anode since the sweeping reduces direct attack on the anode by oxygen or halogen which is formed.

In the case an electrolytic cell arrangement having a horizontal lower anode, the circulation of electrolyte is sufficiently energetic to permit lateral stirring. The gas bubbles of oxygen or halogen are separated by a deflector, formed such that it cannot damage the metal deposit at cathode. This arrangement is particularly advantageous by virtue of the fact that the location of the anode in the bottom of a bath already naturally favours the depolarisation and the reduction of attachment to the anode of oxygen or halogen which is formed there.

In the apparatus of the present invention, the principal feature is the circulation of the electrolyte in a closed circuit around the electrodes. To circulate the electrolyte, the anode(s) and the cathode(s) can be considered to function as electrodes of an electromagnetic conduction pump, so that in arranging the induction conductors creating the electrode current, a magnetic

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field perpendicular to the passage of the current in the electrolyte is created, which forces the electrolyte to circulate between the electrodes, in accordance with Laplace's law. At the end of the electrodes, deflectors separate the electrolyte flow into two flows paths : the first flow path passing the anode and
5 the second flow path passing the cathode. The electrolyte flowing past the anode sweeps the gas bubbles, while the electrolyte flow passing the cathode is loaded with reduced metal.

After the two flows pass the anode and the cathode respectively, these two flow paths rejoin between the electrodes to complete the closed circuit.

10 In a preferred embodiment, an arrangement is used in which an upper anode is used and different deflectors are used according to whether the density of the metal deposited is greater or lesser than the density of the molten salt bath. In fact, these deflectors can be of particularly simple design when used for separating a lighter metal using a cell having a cathode in the upper position or,
15 alternatively, when separating a heavier metal using a cell having a cathode in the lower position. Conversely, in the respective opposing cases, the deflectors can be adapted to facilitate the meeting of the flow paths and thus provides notable reduction to the mechanical wear caused by the circulation of the electrolyte.

20 The apparatus and process of the present invention is herein described with reference to different embodiments. The following description of different embodiments provides further illustrations and examples of the possible adaptations of the process and apparatus to some well known types of industrial electrolysis and can even simplify the known treatment of certain minerals. For
25 example, a first simple embodiment of the apparatus according to the invention can be provided for the electrolysis (ignee) of zinc chloride.

The apparatus and process of the present invention will now be further described with reference to the drawings.

30 Figure 1 is a simplified view of a first embodiment of the apparatus of the present invention. The cell is of a toroidal shape with a vertical axis and the recirculation of the electrolyte is arranged so that the electrolyte flows

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orthogonally through the toroid. The magnetic field is induced by a coil conductor in the form of a toroidal solenoid which is fed in series by current running to the anode or cathode. The anode is comprised of agglomerated graphite in the form of a crown at the bottom of the cell. The cathodes comprise
5 segments of solid crowns adjacent the cover of the cell. The flow deflector is situated on the largest diameter of the toroid, since in that embodiment the direction of the coil of the circuit is the same as between the electrodes, thus providing a centrifugal force in the electrolyte, whereas above the cathodes the circulation is necessarily in the opposite direction. The deflector has a profile
10 suitable for separation of the flows and a system for collection for liquid zinc. Collection tubes collectively guide the liquid zinc towards a circular drain where it may be regularly pumped. The segmentation of the cathode crown is particularly useful for ease of location of the cell cover, but also facilitates the regulation, segment by segment of the distance between the electrodes. The
15 cover is located atop a chloride collector. In addition to the pump for zinc collection, a conduit is provided for feeding of zinc chloride.

Obviously, by an inverting the direction of the current flow in the associated solenoid or, by changing the direction of the winding, the force exerted on the electrolyte can be reversed, causing centripetal flow and the
20 deflector must be moved or relocated accordingly.

Figure 2 is a simplified depiction of an embodiment of a cell adapted for the electrolysis of magnesium chloride. Like the preceding cell, this cell is in the shape of a toroid having a vertical axis and the recirculation of the electrolyte is in a direction orthogonal to the toroid. The magnetic field is induced by a
25 wound conductor in the form of a toroidal solenoid fed in series by the current induced at the cathode. The anode is constituted by graphite agglomerate at the bottom of the cell. The cathode is also in the form of a crown and overhangs the magnetic induction toroid. The flow deflector is reduced to a simple plate interdependent with the cathode. The active surface of the cathode is formed by
30 a grate which leaves a passage for the magnesium so that it can accumulate on the upper part of the hollowed cathode. The profile will be adapted according to

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the direction of circulation be it centrifugal or centripetal. The cover of the cell is mounted on a chloride collector, but also on a basket of carbo-chloride fed continuously with magnesium oxide and a powder of carbon so as to produce the magnesium chloride and cause the liberation of gaseous chlorine. The
5 magnesium pumping apparatus can be any of the many suitable pumping apparatus known in the art and is not shown in the drawings.

In the two preceding descriptions the embodiments of the invention are very similar and differ only slightly to the first embodiment, the metal produced is heavier than the molten salt bath, making it necessary to have a flow separation
10 deflector passing the anode and the cathode, whereas in the second embodiment the metal is lighter than the molten bath, the metal is caught when traversing the cathode, and the deflector is much simplified. This embodiment is generally useful for the electrolysis of lower density salts and metal oxides such as: magnesium, calcium, lithium, sodium, potassium and barium, since all these
15 metals are principally lighter than their salts or their oxides. These relatively low density metals are also the most reductive metals and these are the metals used the most frequently in thermo-metallic reactions for the reduction of metallic oxides. This is why, it becomes particularly advantageous to combine the first inverse electrolytic apparatus having electromagnetic sweeping, with a
20 second reduction cell which uses thermo-metallics or even electrolysis. The combination of the two, permits the production in situ of thermo-metallic reducers. This combination permits not only perceptible improvement of the classic process but also permits the reduction of metal oxides, which are particularly difficult to reduce, such as titanium oxides or zirconium oxides.

25 The figure 3 is a simplified depiction of a first step in the combination of a production cell of a light metal reductor, and of a classic aluminium electrolysis cell. In a first approach, the reductor produced is lithium which is compatible with aluminium. Lithium produced by the swept electrolysis cell is injected at the bottom of the aluminium bath, located adjacent the cathode of the
30 electrolysis cell. Lithium is less dense than the aluminium and tends to rise in the bath and can directly enter into a thermo-metallic reduction reaction with the

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aluminium oxide. The aluminium produced by this reaction tends to accumulate at the bottom while the less dense lithium oxide tends to move towards the surface where it can react with the carbon and form floating liquid lithium carbonate which is removed by overflow. This combination naturally drives a reduction in consumption of graphite of the anodes of the aluminium cells, even if these is replaced by a carbon powder consumption. Through a second approach, we will be able to replace almost the whole cryolite bath by a lithium carbonate bath in which alumina will be in suspension ; by doing so, fluor fumes will be substantially reduced while the graphite consumption will be also be reduced.

Figure 4 shows as indicated earlier arrangement of an electrolytic cell with a sweeping capacity for the production of a reducing alloy, with a metallo-thermal cell still equipped with electrodes. Both cells are intimately linked in order to create only one cell. This cell could be applied to the alumina reduction process but we prefer to describe it hereafter in a titanium dioxide reduction application.

The cell is presented as a cylindertype of tank comprising inside two cores sharing the same vertical axis with in the middle a chlorination reaction at their centre. The lower toroid corresponds to a reducing cell having an alloy reducer. It is constructed in a similar fashion to the cell describe in figure 2. A toroidal solenoid induces a magnetic field perpendicular to the passage of current between the electrodes. The bottom of the tank is comprised by the anode, and the cathode facing it is in the form of a hollowed crown which at the same time performs the function of deflector.

The upper toroid which corresponds to the thermo-metallic cell, comprises a lower half toroid which at the same time, assures the closure of the lower toroid. It is capped by a cover which is also the closure cover of the cell. This upper toroid also functions in the electrolysis tank with a cathode in the form of a crown located at the bottom of the toroidal tank. Several immersed anodes are suspended from the cell cover. The electrodes of the thermo-metallic cell permit the generation of an electric field favourable for the reduction of titanium

dioxide and they are useful for starting off the cell. The cover may also be a collector for carbon-based gas which is generated from the final step of reduction.

5 The lower cell is replenished with a mixture of lithium chloride and sodium chloride. The chlorine electrolysis product is directed towards the reaction zone, while the lithium and sodium are co-products and collected by the hollow cathode, then pumped and injected into the liquid cathode of the upper cell.

10 The upper cell is replenished with lithium carbonate and sodium carbonate which functions as electrolyte and dispersant for the titanium oxide. At the bottom of the upper cell, the liquid cathode is constituted by an alloy bath. The alloy bath is basically heavy metals at lower melting point which is injected to continuously reduce the alloy at the cell base, but this liquid cathode also continuously extracts an alloy rich in titanium. In effect, the alloy reductant of the cathode is oxidised in reducing the titanium oxide which is in suspension in the bath. The oxides formed migrate towards the surface of the bath where, in the presence of carbon, they form liquid carbonates which move to the centre of the cell and the chlorination reaction zone. Accordingly this at the same time, causes the dissociation of carbon-bearing gas and regeneration of the chlorides.

15 If the consumption of the chlorides is theoretically nil or minimal because the chlorides are regenerated in the course of a cycle, and it is necessary to gradually extract the titanium of the cathode alloy to refeed the thermo-metallic cell with titanium dioxide and carbon powder.

20 Figure 5 shows a cell which is similar to the preceding cell, but adapted for the reduction of wolframite. In this double cell, the lower toroid corresponds to the reduction cell and is provide with an alloy reductor. The upper toroid corresponds to the thermo-metallic cell. The lower cell is filled with a mixture of sodium chloride and lithium chloride. The electrolysis produces chlorine which is directed towards the chloride regeneration zone while the sodium and lithium are co-produced and are collected by the hollow cathode, then pumped and injected in the liquid electrode of the upper cell.

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The wolframite is introduced to the heart of the bath of the upper cell. The wolframite is decomposed to tungsten oxides and iron oxides. The tungsten oxide and the sodium oxide form a liquid composition which the essential part of the bath for putting the wolframite into solution. The iron oxide which is in suspension, is reduced. The tungsten oxide is reduce on contact with the electrode.

conversely to the arrangement described in respect of the preceding figure 4, the electrodes to the upper tank are in this embodiment, fed by alternating current and furthermore, an magnetic induction toroid connected in series, creating circulation between the electrodes. This permits better decantation of the bath, facilitates the surface concentration of sodium oxide and also permits elimination of the iron slug. Furthermore, the tungsten penetrates the liquid electrode and concentrates at the lowest point because the tungsten density differs considerably to the densities of the most important metals present.

The sodium oxide returns to the surface, moving progressively In the central regeneration zone, where it reacts in the presence of chlorine and carbon powder, to form chlorine and carbon-bearing gas. The cell cover then become a collector for the carbon-bearing gas.

Thus, this cell permits at the same time, separation of iron and production of a tungsten alloy, for example : an alloy of tungsten-lead-bismuth, an alloy which facilitates the use of tungsten in manufactured products.

A sith embodiment of the apparatus according to the invention allows the direct treatment of concentrates of zinc or lead, without roasting pre-treatment, but with separation of iron impurities present and with condensation of sulphur.

For example, figure 6 shows schematically and partially, a cell for treatment of a concentrated mixture of zinc and lead sulphurs. The cell is of toroidal shape having a vertical axis and the circulation of the electrolyte is arranged so as to have an orthogonal flow in the toroid. The electric field is induced by a rolled conductor in the form of a toroidal solenoid which is fed in series by a current to the anode or the cathode. Like the cell depicted in figure 1, this cell is comprised of an upper cathode and a lower anode in the form of a

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5 crown, but in contrast to figure 1, the diffuser or deflector is now central and the circulation is under the anode. The central diffuser has the function of a basket for dilution of the chloride concentrate. In the upper part of the basket the chloride attacks the lead and zinc sulfates, while in the lower part, the chlorides are diluted in the electrolyte. The sulphur formed by the reaction is condensed by a device which is not depicted in the drawing. The chloride in circulation are reduced by the cathode and are deposited at the bottom of the cell.

10 It is also possible to conceive of a process in which zinc is drawn off, allowing initial concentration of the lead by thermo-metallic processing of zinc, then leading into a first phase of lead production and enrichment of the concentrate of zinc sulphate, and a second phase using the preceding cell for the production of zinc, lead of pre-treatment purity. The improvement of the type already described allow further separation of the iron and impurities which are present.

15 A seventh embodiment of the invention concerns the treatment of dolomite. Dolomite is a double carbonate of magnesium and calcium. Dolomite is an abundant mineral which is can be used similarly or concurrently with magnesium minerals, but it is necessary to perform decarbonation and a separation of calcium oxide. One embodiment according to the present the present invention allows the treatment of dolomite directly after decarbonation, but without the necessity of separating the calcium oxide by pre-treatment. This adaption allows isolation of magnesium and calcium . For example, a first thermo-metallic cell or thermo-calcic cell uses magnesium of decarbonated dolomite. It is possible to use a rotary oven comprising a lead-calcium bath on which the dolomite floats. This oven includes a compartment for condensation of magnesium.

The oven can also be used for the decarbonation which then follows the thermo-calcic treatment. After exhaustion and drawing off of the magnesium, the lime is taken out and the oven is reloaded for a new treatment.

30 The lime formed by the treatment is treated completely, or in part by a sweep cell for calcium production. The calcium produced is alloyed with the

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lead and returned to the rotary oven. This alloy of lead-calcium is returned to the rotary oven, but part can be directly used as a metalo-thermic reductor for other applications or again, be used directly as an alloy for the manufacture of electric accumulator plates.

5 In the previous examples, the cells have been described with a core shape included a coiling up movement in order to create a magnetic field, but they could as well be made in a cylinder shape, while imagining the possibility of opening the core and uncoiling it. In this case, the magnetic field need to be closed by a magnetic breech, but, by twinning two cylinders next to each other,
10 the head breeches of the cylinders will then end up enjoying a very simplified design.

 In a same manner, and according to the same invention, it remains possible to conceive some applications with a vertical or oblique setting of the electrodes, while judiciously planning the electrolyte circulation and the setting
15 of the deflectors.

 Other embodiments can be envisaged to carry out the treatment of ilmenite with iron separation. Numerous other variations can be described to be adapted for the treatment and reduction of different minerals often providing substantial improvements in economy in comparison with existing treatment
20 and technologies of the prior art. The preceding description of embodiments of the apparatus of the mineralogical processes are not exhaustive and further embodiments apparent to the person skilled in the relevant technology are within the scope and spirit of the present invention. The present invention is not to be considered to be limited to the embodiments described.

The claims defining the invention are as follows:

1. An electrolytic cell comprising electrolyte, at least one anode and at least one cathode, characterized as follows : the electrolyte is caused to circulate
5 between the cathode(s) and anode(s) by applying a magnetic field typically perpendicular to the electricity flow in the electrolyte and associated with one or several deflectors which divide the electrolyte flow into two flows paths : the first flow path towards the anode and the second flow path towards the cathode.
2. An electrolytic cell according to claim 1 wherein the two flow paths
10 rejoin between the electrodes to complete a closed circuit.
3. An electrolytic cell according to claim 1 and 2 wherein the cell is of a toroidal shape with a vertical recirculation of the electrolyte is arranged so that the electrolyte flows orthogonally through the toroid and the magnetic field is induced by a conductor in the form of a toroidal solenoid.
- 15 4. An electrolytic cell according to claim 1 and 2 wherein the cell is of a cylindrical shape with a vertical recirculation of the electrolyte is arrange so that the electrolyte flows orthogonally through the cylinder and the magnetic field is induced by a conductor in the form of a cylindrical solenoid, with a magnetic blocs joining the extremity in order to close the magnetic field.
- 20 5. An electrolytic cell according to claim 4 where the cell is associated with another and two magnetic blocks in order to close the magnetic field.
6. An electrolytic cell according to claim 1 wherein the metal is lighter than the molten bath, and the metal is caught when crossing the cathode.
7. An electrolytic cell according to claim 6 wherein the deflector is
25 associated with the cathode.
8. A process using an electrolytic cell according to claim 1 and as described in the patent.

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FIG. 1

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FIG. 2

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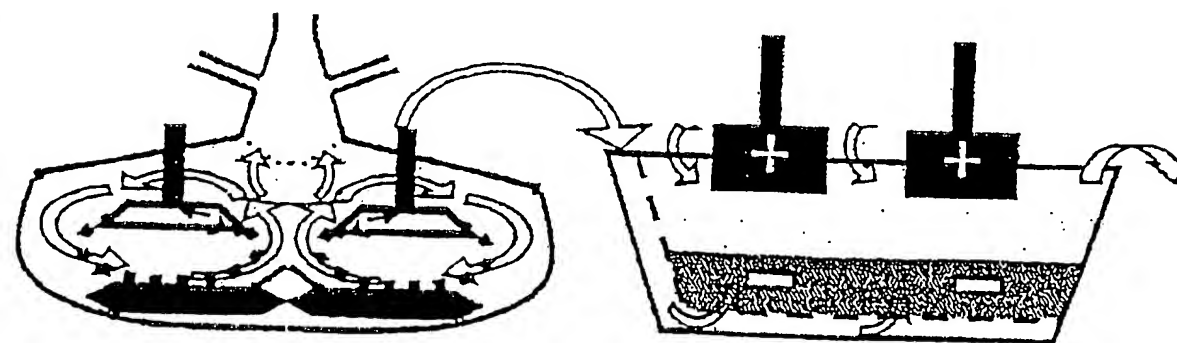
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FIG. 3

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FIG. 4

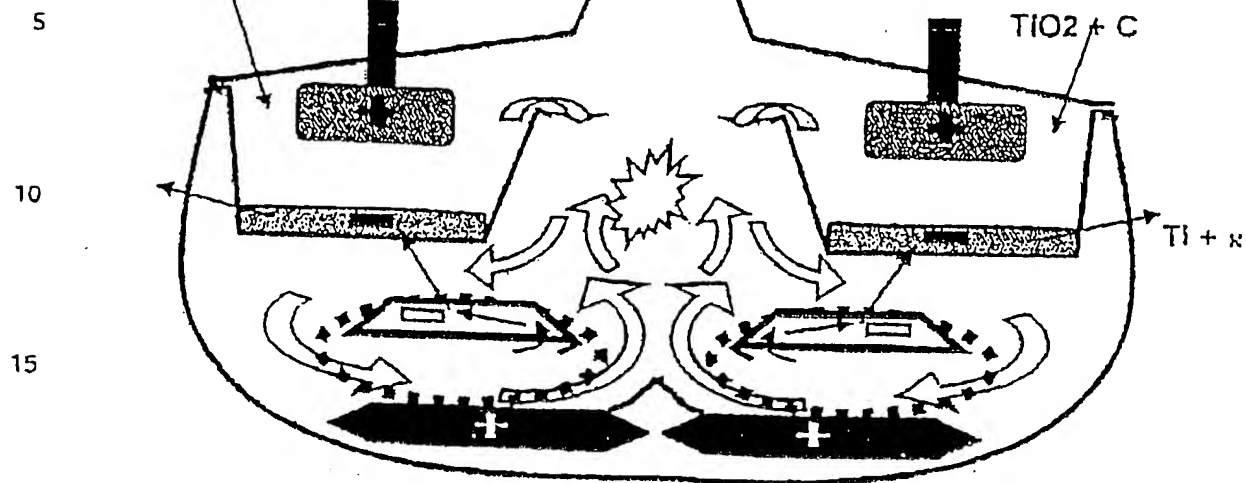


FIG. 5

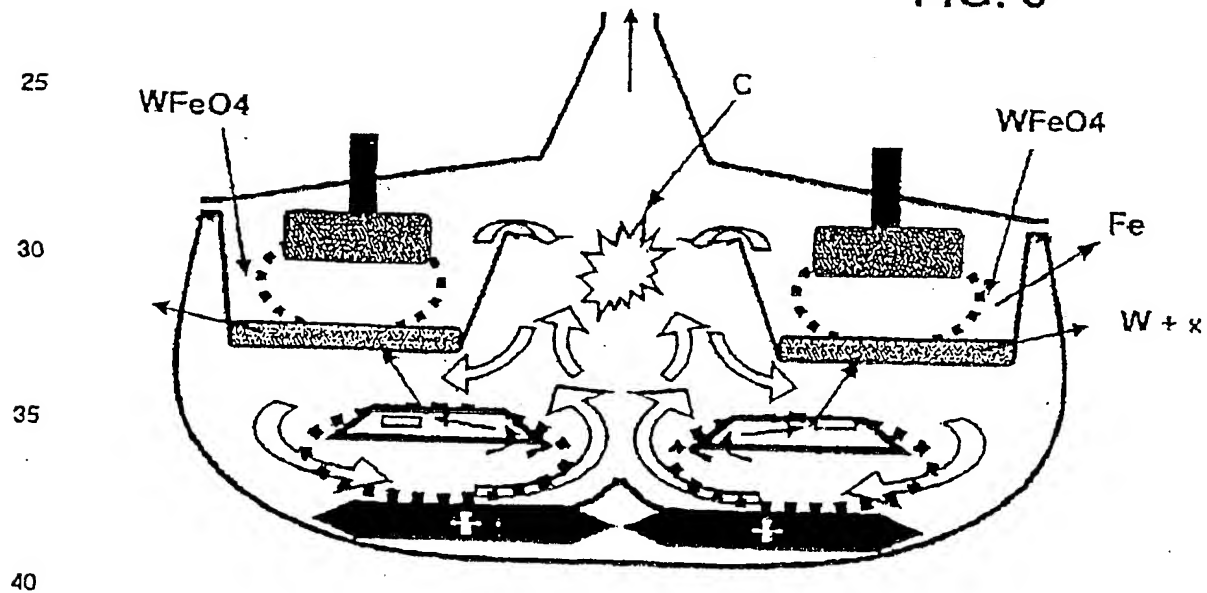
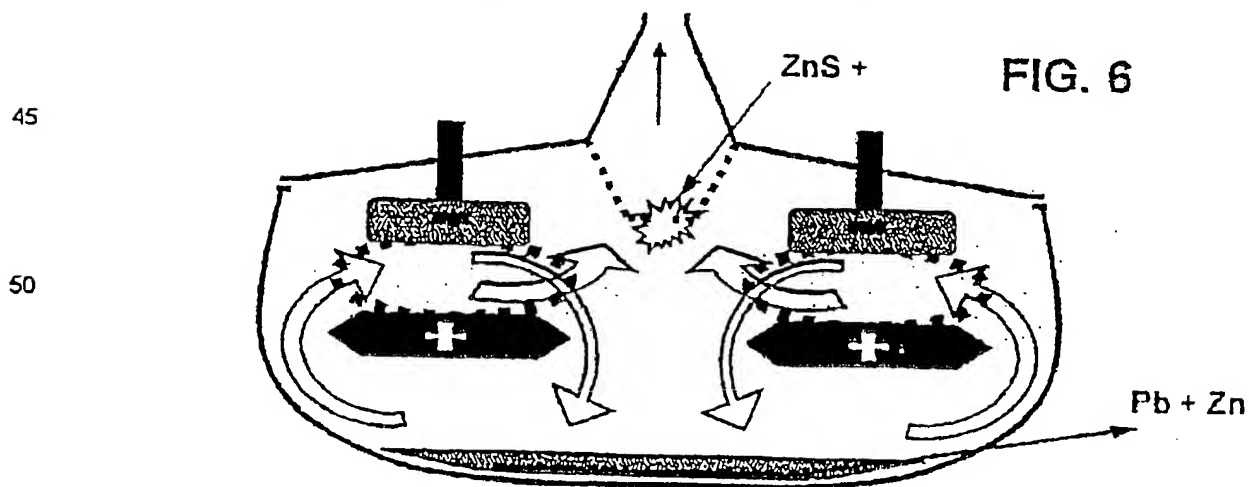


FIG. 6



INTERNATIONAL SEARCH REPORT

International application No.

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A. CLASSIFICATION OF SUBJECT MATTER

Int. Cl. 7: C25C 7/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

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C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 4565748 A (DAHL) 21 January 1986 See Abstract	1-8
A	Patent Abstracts of Japan, P-1509, page 103, JP 04-319699 A (TOSHIBA CORP) 10 November 1992 See Abstract	1-8
A	US 5240569 A (WALDRON) 31 August 1993 See Abstract	1-8

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Date of the actual completion of the international search

22 February 2000

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INTERNATIONAL SEARCH REPORT

International application No.

PCT/AU00/00042

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	FR 2584621 A (VIVES) 16 January 1987 See Abstract	1-8
A	Patent Abstracts Of Japan, C-117, page 108, JP 57-67184 A (MITSUBISHI KEIKINZOKU KOGYO KK) 23 April 1982 See Abstract	1-8

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.
PCT/AU00/00042

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Patent Document Cited in Search Report		Patent Family Member
US	4565748	NONE
JP	04-319699	NONE
US	5240569	NONE
FR	2584621	NONE
JP	57-67184	NONE
		END OF ANNEX

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